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Development of a control system for in-line coagulation in an ultrafiltration process

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Abstract

A control system for the in-line coagulation applied in an ultrafiltration process was developed. The dosing strategy aims to apply a minimal coagulant dose while maintaining desirable process performance by regulating the initial filtration resistance. This is achieved by a feedback controller. It was found that the control system performs well; adaptation to changing conditions is achieved adequately and sufficiently fast. The initial resistance of the last filtration before the chemical cleaning phase can be controlled within an accuracy of approximately 3% (of the total resistance) or 9% (of the fouling resistance). Compared to the current dosing strategy, a significant reduction of coagulant consumption can be achieved.

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1. Introduction

Ultrafiltration plays an important role in the production of potable water. However, (irreversible) membrane fouling is a limitation in the application of this technology. The accumulation of the retained matter on the membrane surface leads to an increase in operating costs, due to an increased energy consumption and the necessity of periodic cleaning. To reduce these operating costs, it is necessary to control the fouling behavior. The fouling behavior is determined by the interaction between the physicochemical feedwater properties, membrane properties and operating conditions.

Natural water is difficult to characterize, since it can contain a large number of different components. However, generally it is found that (irreversible) fouling by natural organic matter (NOM) is enhanced by: decreasing pH [1–5], increasing electrolyte concentration [3,4], increasing NOM molecular weight [11–13], increasing NOM hydrophobicity [7,12] and addition of divalent cations (e.g. Ca^{2+}) [4–11]. Due to the complexity of solution chemistry in natural waters, NOM properties are very

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Regarding the membrane properties, it is observed that irreversible fouling is enhanced, if the membrane is rough [15,16], hydrophobic [5,17–19] or if the pore size is approximately equal to the particle size [20].

A pretreatment method is needed to counter irreversible fouling and operate membrane filtration under economically feasible conditions. Some feedwater pretreatment options for ultrafiltration are coagulation, powdered/granulated activated carbon (PAC/GAC) dosing [23–25] or ozonation [26,27]. Pre-coagulation is a separate process which is followed by conventional flotation or sedimentation. The supernatant is then used as feed for the filtration process. This study, however, is concerned with in-line coagulation, which is the application of a coagulant before membrane filtration without a flotation/sedimentation or pre-filtration step.

Investigation of in-line coagulation has shown that it benefits the performance of the filtration process. A reduction of the hydraulic resistance of the fouling layer can be observed [29–31]. This suggests that either a more permeable cake is formed or the internal membrane surface is better protected from foulants. Furthermore, hydraulic cleaning is more effective [31]. Finally, the permeate quality is better due to enhanced NOM

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and turbidity removal [31,30]. This potentially improves the performance of subsequent process steps (for example RO/NF) and reduces the concentration of disinfection byproduct precursors.

However, application of in-line coagulation does have drawbacks. First, it forms a large portion of the operating costs, due to chemicals consumption and the increased disposal costs of the concentrate stream. Second, coagulant residuals in the permeate, caused by overdosing, reduce the product quality and can lead to issues in downstream processes, for example RO [32]. In some cases it is even observed that dosing of coagulant adversely affects the performance of membrane filtration [6].

Hence, it is desirable to use a good dosing strategy, which applies the minimum addition at which the filtration process shows desired performance. This is different from the conventional optimal coagulant concentration, which is the concentration at which sedimentation yields good results. Compared to the conventional optimum, under-dosing leads to both good filtration properties and good removal of NOM [31]. This observation further motivates the desire for a method for minimal dosing of coagulants.

The current practice is to apply the optimal conventional dose, usually found by jar tests, or to test a number of concentrations in a pilot plant study and selecting the most appropriate one. However, if the dosing is not continuously adapted to seasonal and long term trends in the water composition, alteration of other operating settings and gradual changes in membrane properties, it can be expected that under- or overdosing will occur. In this investigation, this adaptation is achieved by feedback control.

2. Theory

The design of a control system consists of the following steps: definition of control objectives, selection of manipulated and measured variables, selection of the control configuration and finally controller design. An introduction to control systems can be found in Ref. [34]. Subsequently, evaluation of the controller performance is addressed.

2.1. Control objectives

The primary goal of in-line coagulation is stabilization of the filtration process, improvement of permeate quality by enhanced NOM removal is of secondary importance. Here, only stabilization of the filtration sequence is considered. Hence, the amount of fouling that is allowed to accumulate between two chemical cleaning phases needs to be kept within certain bounds.

2.2. Selection of manipulated and controlled variables

To realize the control objective, it should first be quantified. The resistance is a good measure for the amount of fouling present in the system and will serve as controlled variable. The resistance is the sum of the membrane resistance $R_{\rm M}$ and a pro-



Fig. 1. Series of subsequent filtrations and backwashes between two chemical cleaning phases. The line illustrates the development of the resistance profile.

gressively growing fouling resistance $R_{\rm f}$. Darcy's law relates the resistance to the flux J, the transmembrane pressure ΔP and the viscosity μ :

$$R_{\rm M} + R_{\rm f} = \frac{\Delta P}{\mu J} \tag{1}$$

Fig. 1 sketches the resistance during a series of subsequent filtrations and backwashes between two chemical cleaning phases. The initial resistance R_0 is the resistance at the end of a backwash or the start of a filtration phase. The objective, stabilization of the filtration sequence, is to control the final resistance before the chemical cleaning.

In principle, the operating variable that has the most influence on the controlled variable should be chosen as the manipulated variable. The coagulant dose and the filtration flux are the variables that most clearly influence the reversibility. The coagulant dose is chosen, because the reversibility is very sensitive to changes in this dose. Furthermore, the filtration flux is directly related to the produced volume. In many situations the produced volume is determined by external demand or economic considerations, and thus the filtration flux cannot be manipulated freely.

2.3. Control configuration

The control configuration is the structure in which the information flows from the available measurement to the manipulated variable. The interaction between the physicochemical feedwater properties and the membrane surface under the influence of the coagulant dosing and other operating conditions is very complex. A feedback controller is selected because feedback is able to deal with systems of which the behavior is not exactly known. The control configuration, where feedback is used to adapt the coagulant dosing to control the initial resistance, is shown in Fig. 2.

2.4. Controller design

Typically, a feedback controller is used to keep the controlled variable at it an invariant setpoint. However, the control objec-



Fig. 2. Basic feedback configuration.



Fig. 3. Schematic representation of the effect of parameters on the desired initial filtration resistance trajectory.

tive does not require us to keep the amount of fouling constant, providing that the final value is acceptable. The natural shape of a filtration sequence curve shows some accumulation of foulants over the subsequent filtrations (see the dashed line in Fig. 1). To prevent the coagulant dose from being abusively corrected as a result of an infeasible setpoint, an empirical relation is defined, to approximate the natural shape of the filtration sequence curve. This trajectory as a function of the cumulative filtered volume per unit area (V_F) is assumed to be given by

$$R_{0,d}(V_{\rm F}) = R_{\rm M} + \alpha_{\rm i} V_{\rm F} + R_{\rm r} (1 - {\rm e}^{-V_{\rm F}/v_{\rm eq}})$$
(2)

It is assumed that the initial resistance of the first filtration following a chemical cleaning phase is the membrane resistance $R_{\rm M}$. This leaves us three degrees of freedom to define a trajectory, $\alpha_{\rm i}$ is the final slope, $R_{\rm r}$ is gain of the exponential rise and $v_{\rm eq}$ is its characteristic volume. The resulting trajectory can be linear, exponential or a combination. The effect of these parameters on the filtration sequence curve is illustrated in Fig. 3.

Three examples of desired initial resistance trajectories are shown in Fig. 4 and depicted by I, II and III. The circles in the



Fig. 4. Measured (circles) and desired (lines) filtration trajectories as a function of the cumulative filtered volume. The curves are intended to provide a few examples of desired initial filtration resistance curves that may be chosen, II corresponds to the setpoint trajectory in Section 4.2 and III corresponds to the setpoint trajectory in Section 4.3.

figure represent measured values of the initial resistance for a number of subsequent filtration phases. Compared to the measured values, trajectory I might be realized by increasing the coagulant dose, whereas trajectories II and III might be realized by decreasing the coagulant dose.

A deviation from the desired initial filtration resistance trajectory curve indicates that the final value is probably going to deviate from the desired final value as well. Thus, a difference between the desired initial filtration resistance and the measured initial filtration resistance should result in an adaptation of the coagulant dose. The difference between the desired and realized initial filtration resistance is the control error. When the solid line (I) in the figure is chosen as desired trajectory, ε indicates the control error. With n_F the filtration counter, the desired initial resistance trajectory $R_{0,d}(V_F(n_F))$ and the measured initial resistance $R_0(n_F)$, the control error can be defined by the following equation:

$$\varepsilon(n_{\rm F}) = R_0(n_{\rm F}) - R_{0,\rm d}(V_{\rm F}(n_{\rm F})) \tag{3}$$

The controller is the algorithm that determines how the information obtained from the process (the control error) is used to adapt the manipulated variable. Since a trajectory for the initial filtration resistance is tracked, the coagulant dose C_F is adapted one time per filtration, at the moment the initial resistance is estimated. A discrete proportional-integral (PI) controller is used, which can be defined by

$$C_{\rm F}(n_{\rm F}+1) = C_{\rm F,0} + K\left(\varepsilon(n_{\rm F}) + \frac{1}{n_{\rm I}}\sum_{i=1}^{n_{\rm F}}\varepsilon(i)\right) \tag{4}$$

in which $C_{F,0}$ is the initial value of the coagulant dose. In this equation, the proportional part of the control action $(K\varepsilon(n_F))$ expresses that the adaptation of the coagulant dose should be proportional to the control error. The controller gain K should be chosen large enough to ensure that the control action is significant. However, a value that is too large can lead to oscillation or instability of the the controlled system. The integral part of the control action is given by $(K/n_I)\sum_{i=1}^{n_F}\varepsilon(i)$. The magnitude of this term keeps growing until the control error is reduced to zero and thus it is assured that the setpoint is eventually reached. The rate at which this term grows can be tuned by the integration interval n_I . A small value results in a fast response, which may be oscillatory or unstable. On the other hand, a large value results in a smooth and stable response, which may be slow.

When the manipulated variable is constrained, it is more convenient to use the velocity form of Eq. (4), which expresses the change of the manipulated variable $(C_F(n_F + 1) - C_F(n_F))$. Hence, the PI control algorithm may be given in velocity form by

$$C_{\rm F}(n_{\rm F}+1) = C_{\rm F}(n_{\rm F}) + K\left(\left(1+\frac{1}{n_{\rm I}}\right)\varepsilon(n_{\rm F}) - \varepsilon(n_{\rm F}-1)\right)$$
(5)

The bounds can be given by

$$C_{\rm lb} < C_{\rm F}(n_{\rm F}) < C_{\rm ub} \tag{6}$$



Fig. 5. Schematic representation of the pilot-plant scale filtration unit employed in the experiments.

3. Materials and methods

The experiments were performed on a pilot plant scale filtration unit which is schematically shown in Fig. 5. Two Norit-Xiga UFC SXL-225 FSFC modules with a filtration surface of 40 m² each and a cut-off of approximately 200 kDa were used. These consist of hollow fiber porous PES/PVP membranes with an internal diameter 0.8 mm and an effective length of approximately 1.5 m. The internal fiber volume is approximately 161, the additional dead volume of the system is estimated at 81.

Surface water taken from the Twente Canal was used as feed. This water contains, amongst other things: fatty acids, polysaccharides, proteins, high molecular weight organic structures and clay. Most of these substances occur naturally in surface water and originate from, for example, plant material. In addition, some municipal, industrial and agricultural contamination can be found. The feedwater was prefiltered ($200 \,\mu$ m) to prevent too large particles form entering the system. The feedwater was buffered in a continuously refreshed and well stirred feed tank.

Filtration sequences were preceded by a chemical cleaning procedure. This consisted of 20 min soaking in a NaOH solution at pH 11 with an addition of 100 ppm NaOCI. This was followed by 20 min soaking in a HCl solution at a pH of 2.

A commercially available poly-alumina coagulant (Quadrafloc PUS, CAS 990001-02-9, ViVoChem BV) was used. To achieve more accurate dosing the stock solution was diluted by a factor 10. This was done with a mixture of water and hydrochloric acid with the same pH as the stock solution. The coagulant dose was controlled by flow ratio control on a dosing pump (FrC in Fig. 5). The mixing point is just before the filtration pump.

In Fig. 5, the pressure sensors are indicated by PT, the temperature sensors by TT and the flow transmitters by FT. Based on these measurements, the values of the viscosity and resistance are calculated by the control software. The initial value of the filtration resistance is obtained by calculating the average resistance during the first 50 s of the filtration phase. Subsequently, the adaptation of the coagulant dose is calculated and the dosing setpoint is altered.

4. Results

4.1. Open loop experiment

An open loop experiment was performed, the results are shown in Fig. 6. The filtration flux $(J_F = 75 \ l/(m^2h))$, filtered volume $(v_F = 0.025 \ m^3/m^2)$, backwash flux $(J_B = 250 \ l/(m^2h))$ and backwash duration $(t_B = 60 \ s)$ were all kept constant. The top graph shows step-changes that were made in the coagulant dose, whereas the bottom graph shows the effect of these changes on the initial resistances. It can be seen that by



Fig. 6. Top: coagulant dose vs. filtration number, bottom: initial resistance vs. filtration number. Other settings were $J_F = 75 \text{ l/}(\text{m}^2 \text{ h})$, $t_F = 15 \text{ min}$, $J_B = 250 \text{ l/}(\text{m}^2 \text{ h})$, $t_B = 60 \text{ s}$.

lowering the dose the initial resistance increases and vice versa and that these effects occur within a few filtration phases. This confirms that the coagulant dose is a suitable control variable.

Looking at Fig. 6 in more detail, it can be seen that during the first 81 filtrations at a dose of 1.0 ppm, the resistance reached a stable value of 7.45×10^{11} m⁻¹. After a subsequent period of 83 filtrations at 0.5 ppm, the dose was increased again to 1.0 ppm. This resulted in a stable resistance of 9.60×10^{11} m⁻¹. From this it is concluded that the effect of decreasing the dose is not necessarily reversible by increasing the dose by the same amount.

A system is called controllable if by using admissible inputs it is possible to steer the system from any initial to any final state. Since irreversible fouling cannot be removed, it is by definition not possible to reach any state from any given initial state. Controllability is an important property of systems to be controlled and the intrinsic lack of this property has an important consequence: the setpoint trajectory needs to be chosen with care to ensure the controller is able to track the desired trajectory. If it is attempted to impose an infeasible setpoint, the controlled system can be unstable.

4.2. Controller tuning

From Fig. 6 it is estimated that a change in coagulant dose of 0.5 ppm results in a resistance change equal to approximately 4×10^{11} m⁻¹, this is about the same for both increasing and decreasing the concentration. Based on this process gain, a suitable gain of the coagulant controller should be approximately 1×10^{-12} ppm m. The number of filtrations needed to achieve most of the change is roughly estimated to be 20. The reaction to an increase in the coagulant dose is much faster (approximately five filtrations). Based on these numbers, the integration interval of the coagulant controller should be chosen equal to approximately 10 filtrations.

The selection of the desired initial resistance trajectory parameters is in principle arbitrary. Thus, a wide variety of trajectories may be realizable, which can be selected to satisfy certain operational objectives. However, the selection of a good or optimal trajectory is beyond the scope of this study. In consideration of the controllability, the parameters were chosen such that the desired trajectory seems feasible compared to the available measured trajectory. It is defined by Eq. (2) with $\alpha_i = 0 \text{ m}^{-2}$, $R_r = 3 \times 10^{11} \text{ m}^{-1}$ and $v_{eq} = 0.1 \text{ m}$. The resulting curve is plotted as a dashed line (II) in Fig. 4.

The controller was implemented in the control software of a pilot plant. Its performance is evaluated by applying the control to a sequence of filtrations. The filtration flux ($J_F = 75 \ l/(m^2 h)$), the filtration duration ($t_F = 600 \ s$), the backwash flux ($J_B = 250 \ l/(m^2 h)$) and the backwash duration ($t_B = 60 \ s$) where all kept constant. The initial coagulant dose was taken as 0 ppm. The result is shown in Fig. 7. The top graph shows the desired and measured resistance and the bottom graph shows the coagulant dose.

In the first hour (six filtrations) the measured initial resistance is lower than the desired initial resistance. The controller should



in that case decrease the coagulant dose, however, since it is already at its lower bound of 0 ppm, it is maintained at this level. After the first hour the initial resistance keeps increasing and it becomes clear that filtration with no coagulant dosing leads to an unstable sequence. To compensate for the increasing resistance, the controller keeps increasing the coagulant dose, until after about 6 h the initial resistance starts decreasing. After approximately 8 h the initial resistance reaches its setpoint. From this point onwards only small variations in the coagulant dose occur, which are used to counter small deviations in the initial resistance.

From Fig. 7 it is concluded that the controller performs well and that no adjustments of the control parameters are necessary.

4.3. Controller evaluation

The performance of the controller was also tested on a number (40) of filtration sequences. Different values for the filtration flux and filtered volume were applied (see Table 1). The desired initial filtration resistance trajectory was defined by Eq. (2), with $\alpha_i = 1.0 \times 10^{11} \text{ m}^{-2}$, $R_r = 3 \times 10^{11} \text{ m}^{-1}$ and $v_{eq} = 0.1 \text{ m}$. The corresponding curve is indicated by a dashed line (III) in Fig. 4. The backwash flux ($J_B = 250 \text{ l/(m^2 h)}$) and the backwash duration ($t_B = 45 \text{ s}$) were kept constant. For surface water with a turbidity in the range of 5–15 NTU typically a coagulant dose of 2 ppm would be used. This was chosen as initial dose. The results are shown in Fig. 8. The top graph shows the measured and desired initial resistance, the middle graph shows the control error and the bottom graph shows the coagulant dose.

Table 1 Values of the filtration flux and filtration duration used in Fig. 8

	а	b	c	d	e	f	g	h	i	j	k
$\frac{J_{\rm F} (l/(m^2 h))}{t_{\rm F} (\min)}$	75	25	50	100	75	75	100	75	75	75	75
	20	60	30	15	10	20	15	60	20	6	20





Fig. 8. Performance of the coagulant controller on a series of chemical cleaning cycles.

It can be seen that due to the high initial dosing, the measured initial resistances are well below the desired trajectory. Consequently the dose is lowered. At the third chemical cleaning cycle, the desired trajectory is reached and the coagulant dosing reaches a steady state. It can be seen that a change in the filtration flux leads to a change in the measured resistance (e.g. going from a to b in Fig. 8). This is caused by the height difference between the pressure sensors, which is not compensated for in the control software. This offset does not influence the results in filtration sequences in which the filtration flux is constant.

The average control error of the initial resistance of the final filtration phase is approximately 9% of the fouling resistance or 3% of the total resistance. Due to an observed overshoot at the beginning of the sequences and the changes in operational settings the average control error evaluated over the entire trajectory is larger (20% and 7%).

4.4. Conclusion

A dosing strategy for in-line coagulation with ultrafiltration was developed. The main objective of this control strategy was to stabilize the sequence of filtrations. The designed controller is able to fulfill this; the initial resistance of the last filtration before the chemical cleaning phase can be controlled within an accuracy of approximately 3% (of the total resistance) or 9% (of the fouling resistance). It was furthermore found that the controller is able to adapt to changes in operating settings. Compared to the current coagulant dosing strategy a large reduction in coagulant consumption can be achieved.

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Nomenclature

C_{F}	coagulant dose (ppm)						
$C_{\mathrm{F,0}}$	initial value of the coagulant dose (ppm)						
$C_{\rm lb}$	lower bound for the coagulant dose (ppm)						
$C_{\rm ub}$	upper bound for the coagulant dose (ppm)						
J	flux (m/s)						
J_{B}	average backwash flux $(l/(m^2 h))$						
$J_{ m F}$	average filtration flux $(1/(m^2 h))$						
Κ	controller gain (ppm m)						
$n_{\rm F}$	filtration number						
$n_{\rm I}$	integration interval						
ΔP	transmembrane pressure (Pa)						
R_{f}	fouling layer resistance (m^{-1})						
R _M	membrane resistance (m^{-1})						
$R_{\rm r}$	initial rise of the desired initial filtration resistance						
	trajectory (m^{-1})						
R_0	initial filtration resistance (m^{-1})						
$R_{0,d}$	desired initial filtration resistance (m^{-1})						
tB	backwash duration (s)						
$t_{\rm F}$	filtration duration (min)						
v_{eq}	characteristic volume of the desired initial filtra-						
	tion resistance trajectory (m)						
$V_{ m F}$	cumulative filtered volume per area (m)						
Greek symbols							
α_{i}	final slope of the desired initial filtration resis-						
	tance trajectory (m^{-2})						

 ε control error (m⁻¹)

 μ viscosity (Pa s)

References

- K.L. Jones, C.R. O'Melia, Protein and humic acid adsorption onto hydrophilic membrane surfaces: effects of pH and ionic strength, J. Membr. Sci. 165 (2000) 31–46.
- [2] K.L. Jones, C.R. O'Melia, Ultrafiltration of protein and humic substances: effect of solution chemistry on fouling and flux decline, J. Membr. Sci. 193 (2001) 163–173.
- [3] M. Kulovaara, S. Metsämuuronen, M. Nyström, Effects of aquatic humic substances on a hydrophobic ultrafiltration membrane, Chemosphere 15 (1999) 3485–3496.
- [4] S. Hong, M. Elimelech, Chemical and physical aspects of natural organic matter (NOM) fouling of nanofiltration membranes, J. Membr. Sci. 132 (1997) 159–181.
- [5] M. Kabsch-Kurbutowicz, K. Majewska-Nowak, T. Winnicki, Analysis of membrane fouling in the treatment of water solutions containing humic acids and mineral salts, Desalination 126 (1999) 179–185.
- [6] A. Maartens, P. Swart, E.P. Jacobs, Feedwater pre-treatment: methods to reduce membrane fouling by natural organic matter, J. Membr. Sci. 163 (1999) 51–62.
- [7] A.I. Schäfer, A.G. Fane, T.D. Waite, Nanofiltration of natural organic matter: removal, fouling and the influence of multivalent ions, Desalination 118 (1998) 109–122.
- [8] A.I. Schäfer, U. Schwicker, M.M. Fischer, A.G. Fane, T.D. Waite, Microfiltration of colloids and natural organic matter, J. Membr. Sci. 171 (2000) 151–172.
- [9] S. Yoon, C. Lee, K. Kim, A.G. Fane, Effect of calcium ion in the fouling of nanofilter by humic acid in drinking water production, Water Res. 32 (1998) 2180–2186.

- [10] A. Seidel, M. Elimelech, Coupling between chemical and physical interactions in natural organic matter (NOM) fouling of nanofiltration membranes: implications for fouling control, J. Membr. Sci. 203 (2002) 245– 255.
- [11] A.E. Childress, M. Elimelech, Effect of solution chemistry on the surface charge of polymeric reverse osmosis and nanofiltration membranes, J. Membr. Sci. 119 (1996) 253–286.
- [12] E. Aoustin, A.I. Schäfer, A.G. Fane, T.D. Waite, Ultrafiltration of natural organic matter, Sep./Purif. Technol. 22–23 (2001) 63–78.
- [13] C. Lin, Y. Huang, O.J. Hao, Ultrafiltration processes for removing humic substances: effect of molecular weight fractions and PAC treatment, Water Res. 33 (1999) 1252–1264.
- [14] M.J. Scott, M.N. Jones, C. Woof, B. Simon, E. Tipping, The molecular properties of humic substances isolated from a UK upland peat system, Environ. Int. 27 (2001) 449–462.
- [15] M. Elimelech, X. Zhu, A.E. Childress, S. Hong, Role of membrane surface morphology in colloidal fouling of cellulose acetate and composite aromatic polyamide reverse osmosis membranes, J. Membr. Sci. 127 (1997) 101–109.
- [16] E.M. Vrijenhoek, S. Hong, M. Elimelech, Influence of membrane surface properties on initial rate of colloidal fouling of reverse osmosis and nanofiltration membranes, J. Membr. Sci. 188 (2001) 115– 128.
- [17] W.R. Bowen, T.A. Doneva, H. Yin, Separation of humic acid from a model surface water with PSU/SPEEK blend UF/NF membranes, J. Membr. Sci. 206 (2002) 417–429.
- [18] C. Jucker, M.M. Clark, Adsorption of aquatic humic substances on hydrophobic ultrafiltration membranes, J. Membr. Sci. 97 (1994) 37–52.

- [19] A. Maartens, P. Swart, E.P. Jacobs, Membrane pretreatment: a method for reducing fouling by natural organic matter, J. Colloid Interf. Sci. 221 (2000) 137–142.
- [20] A.I. Schäfer, A.G. Fane, T.D. Waite, Fouling effects on rejection in the membrane filtration of natural waters, J. Membr. Sci. 131 (2000) 37–52.
- [23] C. Campos, B.J. Marifias, V.L. Snoeyink, I. Baudin, J.M. Laîné, Adsorption of trace organic compounds in CRISTAIL processes, Desalination 117 (1998) 265–271.
- [24] Y. Matsui, A. Yuasa, K. Ariga, Removal of a synthetic organic chemical by PAC-UF systems. I. Theory and modelling, Water Res. 35 (2001) 455–463.
- [25] Y. Matsui, F. Coals, A. Yuasa, Removal of a synthetic organic chemical by PAC-UF systems. II. Model application, Water Res. 35 (2001) 464–470.
- [26] S. Takizawaa, K. Fujitab, K.H. Soob, Membrane fouling decrease by microfiltration with ozone scrubbing, Desalination 106 (1996) 423–426.
- [27] B. Schlichter, V. Mavrov, H. Chmiel, Study of a hybrid process combining ozonation and membrane filtration–filtration model solutions, Desalination 156 (2003) 257–265.
- [29] S.J. Judd, P. Hillis, Optimization of combined coagulation and microfiltration for water treatment, Water Res. 35 (2001) 2895–2904.
- [30] C. Guigui, J.C. Rouch, L. Durand-Bourlier, V. Bonnelye, P. Aptel, Impact of coagulation conditions on the in-line coagulation/UF process for drinking water production, Desalination 147 (2002) 95–100.
- [31] K.Y. Choi, B.A. Dempsey, In-line coagulation with low-pressure membrane filtration, Water Res. 38 (2004) 4271–4281.
- [32] C.J. Gabelich, T.I. Yun, B.M. Coffey, I.H. Suffet, Effects of aluminium sulfate and ferric chloride coagulant residuals on polyamide membrane performance, Desalination 150 (2002) 15–30.
- [34] G. Stephanopoulos, Chemical Process Control, Prentice Hall, 1984.